

Thermoluminescence and Electron Spin Resonance Investigations of Minerals for the Detection of Irradiated Foods

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Thermoluminescence (TL) measurements are used to detect irradiated foods. They are performed by investigating the minerals that contaminate certain products. Pure quartz, feldspars, and mineral mixtures, which are to be expected in foods, were examined for their radiation sensitivity, dose response, stability of signals, and changes of sensitivity induced by prior heating and prior irradiation. The sensitivity of all samples investigated varied over a range of 3 logs. Dose dependency and signal stability as a function of storage temperature and light exposure were similar for all samples. Prior heating and irradiation treatments significantly affected the sample sensitivity, which might be a reason for the high variability of results known from routine food control. The composition of the mixtures was analyzed by X-ray diffraction spectroscopy. It was found that almost all samples contained a significant amount of quartz in addition to minor amounts of feldspars. ESR measurements were performed to attempt to detect and identify the trapped charges responsible for TL. However, the differences in signal stability, dose dependency, and sensitivity imply that the ESR signals do not correlate with the presence of these charges.

Keywords: *Thermoluminescence; electron spin resonance; minerals; food irradiation*

INTRODUCTION

Food irradiation is a means to enhance food safety by the deactivation of pathogen microorganisms and to reduce food losses caused by microbiological contaminants, infestation by pests, sprouting, and ripening. For example, the irradiation of shellfish and some sensitive fruit such as strawberries, mangoes, and papayas as well as of dried fruit and liquid egg products is important for commercial use. In various countries, food irradiation is permitted but there are regulations on which kinds of products are eligible for treatment and limits on maximum dose. In some countries, such food treatment is prohibited or subject to labeling requirements. Therefore, analytical control methods are needed for the detection of irradiation.

In recent years, much research has been carried out to find reliable detection methods. Thermoluminescence (TL) is a method by which charges formed in and trapped by the medium being irradiated are released upon thermal excitation and detected by the light emitted. It was first applied by Heide and Bögl (1984) to detect the irradiation treatment of spices and herbs by measuring whole samples. The finding of the mineral contaminants to be the origin of TL in such samples has led to the development of a mechanical method to isolate these particles from the surface of food samples by utilizing the different densities of minerals and organic components (Sanderson et al., 1989a,b). Autio and Pinnioja (1993) applied TL to the examination of minerals isolated from the intestines of crustaceans, which extended the applicability of the method. Be-

cause of the enormous variability of the mass-normalized TL signal intensities, it was realized that subsequent irradiation with a known dose and a second measurement could serve as a better normalization procedure of the signal, so that, in turn, the detection limits and the reliability of the results were largely improved (Sanderson et al., 1989b). Various experiments on the TL behavior of minerals isolated from foods were reported by Autio and Pinnioja (1990), Schreiber et al. (1993a, 1994), and Sanderson et al. (1996). Several intercomparison studies on herbs, spices, fruits, vegetables, and shellfish (Schreiber et al., 1993b, 1995; Sanderson et al., 1992) confirmed the suitability of the method. The method is recognized as a European standard (EN 1788, 1997).

The experiments described in this paper were performed on samples taken from agricultural areas and from beaches. These mineral mixtures would be expected on foods such as fruits, vegetables, and shellfish. For comparison, some pure quartz and feldspar samples were analyzed. The aim was to find the principal characteristics of sensitivity, signal stability, dose response, and dependence of the sensitivity on prior heating and irradiation treatments to draw conclusions regarding the degree to which these influences might affect the TL results in routine food control. X-ray diffraction spectroscopy served as a means to analyze the composition of the mineral mixtures. Furthermore, experiments were undertaken to determine whether the signals detected by ESR are the same responsible for TL.

MATERIALS AND METHODS

Preparation of Samples. Mineral samples from the surface of 18 agricultural areas in Germany (sample codes FS 01–FS 16), Tenerife (sample code FS 17), and Australia

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(sample code FS 18) as well as from 12 beaches from all over the world (sample codes QS 01–QS 12) were analyzed. Besides these natural samples, four standard samples of pure minerals were under investigation: two quartz samples (Merck quartz and Merck sea sand—both Merck index 11 8440) and two feldspar samples [standard feldspar F1 obtained from the Analytical Quality Control Service (AQCS) of the IAEA (Vienna, Austria) and a microcline Amberger feldspar from a pottery production in Germany].

The 30 natural mineral samples were sieved (resulting granulometry $<500\ \mu\text{m}$). The organic components were removed by density separation in a sodium polytungstate solution of 2 g/mL. Acid-soluble contents of calcium carbonate were removed by 6 M HCl. The four standard samples having a grain size of 125–250 μm were selected by sieving. The irradiation was carried out with a cobalt-60 source at a dose rate of 8 Gy/min. In some cases, the reirradiation was accomplished by using a strontium-90 β -radiation source with a dose rate of 1.7 Gy/min.

TL Analyses. TL glow curves were recorded on a Risø TL reader (Risø, Roskilde, Denmark) working in the single photon counting mode (Bøtter-Jensen, 1988). A linear heating rate of 6 °C/s with an isothermal step of 5 s at 70 °C and a maximum temperature of 500 °C was applied. The integration of the glow curves obtained was carried out between 70 and 500 °C and, additionally, in four subranges of 70–200, 200–300, 300–400, and 400–500 °C. To make the temperature scale comparable to that of other TL readers, it should be pointed out that the maximum of the LiF (TLD 100) glow curve in this TL reader appears at 295 °C under the conditions applied. In the literature, the peak temperature is reported to be ~210–230 °C (McKeever, 1988; Mahesh et al., 1989). The signal intensities of the first measurements were normalized by a reirradiation with doses as stated under Results and Discussion and a following glowing. Results are given as the TL ratios between the first and the second intensities measured. The means and standard deviations were calculated for logarithmic distribution.

To examine the changes in sensitivity caused by irradiation and by extended heating periods, the following measurement procedure was carried out. Samples QS 01–QS 06, FS 01–FS 06, and the four standard samples were irradiated (strontium-90 source, 25 Gy) and measured three times and then treated with the radiation or heat procedure under investigation (see Results and Discussion). Afterward, the samples were measured once to eliminate the signals induced by this radiation or heat procedure. They were then irradiated and measured again three times under the same conditions as before. The differences between the means of the three measurements before and after the treatments are discussed. Values were calculated as percentages of these means to enable a direct comparison between the integration areas. Heat treatments were carried out for 10 min at temperatures between 200 and 500 °C at intervals of 50 °C.

ESR Analyses. The qualitative ESR spectra to obtain the exact g values were measured with a Bruker X-Band ESP300e (Bruker, Rheinstetten, Germany) research instrument. Because of the higher constancy of signal intensities, the quantitative results were determined from measurements with a Bruker EMS104 routine instrument. The measurement parameters were the following: sweep width, 5 mT; modulation amplitude, 0.06 mT (ESPI00e) or 0.14 mT (EMS104); microwave power, 0.6 mW (ESPI00e) or 1 mW (EMS104); microwave frequency, ≈ 9.8 GHz.

X-ray Diffraction Analyses. X-ray diffraction measurements were carried out as powder spectra in the range from 3 to 73 grade at a rate of 1.2 grades/min. The spectra were analyzed by comparison with literature values (Bayliss et al., 1980).

RESULTS AND DISCUSSION

TL Glow Curve Shape and Sensitivity. The shape of the TL glow curves of a part of the irradiated

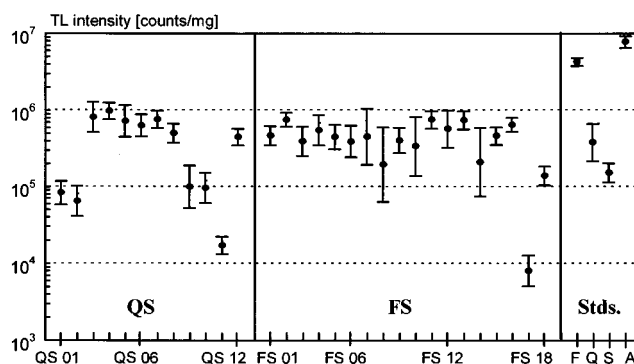


Figure 1. TL sensitivity of mineral (QS, FS in ascending order) and standard samples (standards: F, standard feldspar F1; Q, Merck quartz; S, Merck sea sand; A, Amberger feldspar): means and standard deviations of six disks of each sample, dose 1 kGy, integration area 70–500 °C.

mineral samples was dominated by one peak similar in shape to that of pure feldspars, whereas others showed two to four overlapping peaks comparable to those of quartz glow curves. The shape was not dependent on the origin of the samples. In some cases, portions of material of the same origin exhibited different glow curve shapes.

The mass-normalized TL sensitivity of all analyzed irradiated samples (dose = 1 kGy, measurements 2 days after irradiation) varied over 3 logs. As shown in Figure 1, most of the samples exhibited luminescence in the range of 10^5 – 10^6 counts/mg with maximum standard deviations over one decade within one mineral sample. This represents the high variability of TL even if the materials are of the same origin. Two samples (QS 11 and FS 17) had a very low sensitivity. The reason is that these samples originated from a volcanic place (Tenerife) and did not contain significant amounts of crystalline matter, which was revealed by the X-ray diffraction spectroscopy. The very small TL sensitivity implies that irradiated samples like these might not be detectable by TL analysis in food control. Five of the other mineral samples yielded TL intensities of $\sim 10^5$ counts/mg, which is half a decade lower than the other samples. When compared to the standard samples, it became obvious that pure quartz had a TL sensitivity in the same range, whereas the two pure feldspars exhibited much higher intensities.

X-ray Diffraction Spectroscopy. The X-ray diffraction spectra of the mineral samples showed that all samples (except QS 11 and FS 17) contained quartz as the major component. Besides the specific spectral pattern of quartz, small peaks of feldspars were found, representing smaller contents of material of this mineral group (Figure 2). The five samples with a relatively low TL sensitivity contained the smallest amounts of feldspars as compared to the other samples. It seems to be certain that despite the high quantity of quartz in all samples, the TL sensitivity was dominated by the feldspars. The TL characteristics of quartz influenced the TL behavior of a mixture only if almost no feldspars were present. Identification of the individual feldspars was not possible because of the overlapping X-ray diffraction peaks.

ESR Spectra. Figure 3a demonstrates a typical ESR spectrum of an irradiated mineral sample. Almost all samples of environmental origin yielded spectra of this type with major lines of $g = 2.0006$ and 1.9975 and some minor lines of about $g = 2.0020 \pm 0.0010$. The spectra

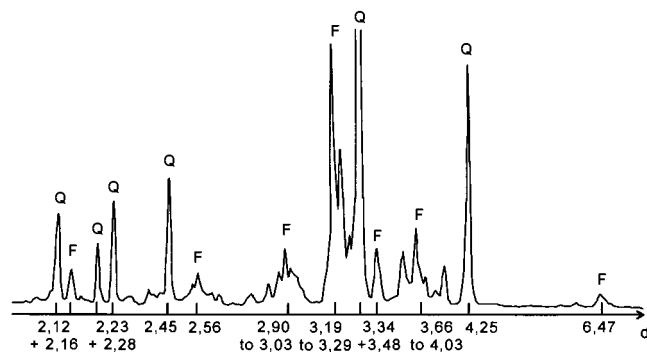


Figure 2. X-ray diffraction spectrum of the mineral sample QS 12. Quartz-specific peaks are called Q; feldspar-specific ranges are called F.

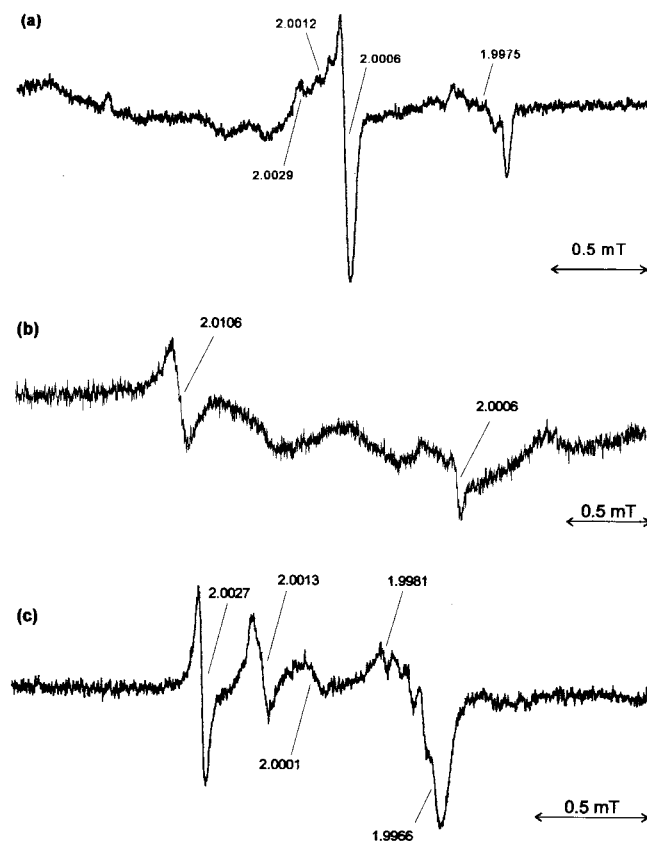


Figure 3. ESR spectra of the irradiated mineral sample QS 01 (a), feldspar (b), and quartz (c) with g values of the lines. Irradiation dose = 1 kGy.

of the standard feldspar samples exhibited only two minor lines with $g = 2.0006$ and 2.0106 (Figure 3b). The standard quartz samples yielded spectra with more intensive lines with $g = 2.0027$, 2.0013 , and 2.0001 and a complex signal with $g = 1.9981$ to 1.9966 consisting of at least five lines (Figure 3c). Thus, it is concluded that the origin of the line $g = 2.0006$ in mineral samples might be due to feldspar, whereas the other lines observed were produced by the quartz contained.

Natural mineral samples yielded quite the same ESR spectra as did the irradiated ones. Only the signal with $g = 1.9975$, being very small among the spectra of natural samples, increased significantly by irradiation. To confirm the assumption that the measured ESR lines were induced by irradiation, the samples were heated ($500\text{ }^{\circ}\text{C}$, 15 min). After this treatment, all lines disappeared, except a small signal with $g = 2.0029$. A subsequent irradiation produced all lines as detected

Table 1. TL Signal Losses (Percent) of Irradiated Mineral Mixtures (1 kGy) under Various Storage Conditions

storage time (days)	integration area ($^{\circ}\text{C}$)	storage condition					
		$-18\text{ }^{\circ}\text{C}$ (freezer)		$4\text{ }^{\circ}\text{C}$ (refrigerator)		room temperature	
		dark	light	dark	light	dark	light
10	70–200	10	90	55	60	95	95
	200–300	10	85	30	40	50	70
	300–400	<10	60	10	20	10	45
	400–500	<10	30	<10	<10	11	25
100	70–200	30	>99	>95	>95	>98	>99
	200–300	20	>99	40	85	70	85
	300–400	10	75	10	70	20	65
	400–500	<10	50	<10	40	10	40

^a Light sources: 20 W halogen lamp, distance of 25 cm, continuous illumination at $-18\text{ }^{\circ}\text{C}$; natural daylight at $4\text{ }^{\circ}\text{C}$ and room temperature. The given amounts are estimations that have to be expected for normalized TL intensities. The initial losses within the first 2 days after the irradiation were not included because in practice the measurements are usually not carried out in this time.

before. Thus, it was proven that the lines described above were irradiation-specific. Although the lines are not heat-stable, no correlation between mass-normalized TL and ESR sensitivities could be established, indicating that TL and ESR signals are due to different trapped charges. The lines in natural samples had certainly been caused by the natural long-time radiation in the environment.

Signal Stability of TL and ESR Signals. To examine storage effects under commercial conditions, six mineral samples of the QS group (QS 01–QS 06) and six samples of the FS group (FS 01–FS 06) were irradiated with a dose of 1 kGy and stored at $-18\text{ }^{\circ}\text{C}$ (freezer), at $4\text{ }^{\circ}\text{C}$ (refrigerator), and at room temperature with and without exposure to light. Measurements were carried out eight times within a storage period of up to 120 days. The first measurements (intensities are set as 100% in the following discussion) were performed 5 h after the irradiation. For TL, pairs of disks were measured each time just once, whereas for ESR, the same samples were measured over the entire period because of the nondestructive nature of the ESR procedure. For reirradiation of the TL samples, a strontium-90 source was used with a constant dose of $\sim 50\text{ Gy}$. The second glow curves were recorded 35 h afterward.

The TL signals of all investigated samples showed similar stability behaviors. Higher degrees of signal stability were observed with lower storage temperatures and increased integration temperature. Light exposure reduced signal intensities significantly at all temperatures applied. Intensive artificial lighting at $-18\text{ }^{\circ}\text{C}$ (20 W halogen lamp, distance of 25 cm) resulted in a larger decrease than that produced by daylight at $4\text{ }^{\circ}\text{C}$ and at room temperature. In Table 1, the TL signal losses caused by storage, which have to be expected in routine food control, are summarized. These results are important in the discussion about threshold values for the detection of irradiated foods and about dose estimations for unknown samples.

Figure 4 demonstrates the signal stability at room temperature with daylight exposure. Horizontal lines show the detection limits in every integration area determined by six parallel measurements of each unirradiated mineral sample with the same reirradiation procedure as in the storage experiments. It can be seen

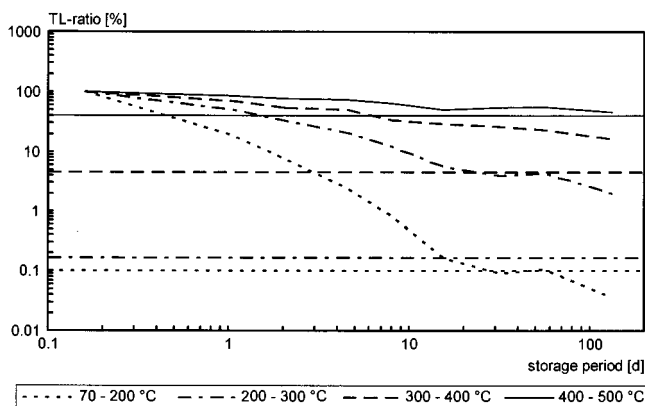


Figure 4. TL signal stability in the four integration areas at room temperature with daylight exposure: means of 12 different mineral samples (duplicate analyses of each). The TL ratio of first to second glow is calculated as percentage of the first measurement 5 h after the irradiation (1 kGy) to make possible a direct comparison between the integration areas. Standard deviations amounted to $\sim 30\%$. Horizontal lines indicate the detection limits in every integration area, determined by the measurements of six disks of each natural mineral sample (mean plus 3 times the standard deviation) with the same reirradiation procedure.

that the mean signal intensity in the lowest integration area (70–200 °C) within 30 days decreased to below the detection limit while the signals in the higher integration areas did not. Because of the high detection limits in the higher integration areas caused by natural radiation, the range of 200–300 °C is the optimum integration area for the detection of intentionally irradiated samples. In Figure 4, this is indicated by the largest difference between the detection limit and the signal intensity after extended storage periods. Although losses, shown in Table 1, seem to be high, the irradiation treatment of all samples was detectable in the integration range 200–300 °C during the whole storage period investigated. This result confirms former findings in interlaboratory studies reported by Schreiber et al. (1993b, 1994, 1995).

By investigating the standard samples of feldspar F1, Merck quartz, and Merck sea sand, it was found that the fading was similar when light had been avoided. Light exposure resulted in a much faster loss of signal intensities in feldspar as compared to quartz and sea sand even in the higher integration areas. This implies that different traps with different photosensitivities cause TL in quartz and in feldspars. In the natural mineral samples, mixtures of both mineral groups are present so that the fading characteristics overlap.

By analyzing the ESR spectra, it was observed that all signals were stable over a period of 100 days within a deviation range of $\pm 20\%$.

Dose Dependency of TL and ESR Signals. To estimate the dose dependency of TL and ESR signals, mineral samples QS 01–QS 06, FS 01–FS 06, and the four standard minerals were irradiated with eight different doses between 0.05 and 10 kGy. Three parallel TL measurements of each sample and dose were carried out 2 weeks after irradiation. TL intensities were normalized by a second glowing 2 weeks after the reirradiation by a dose of 2 kGy (± 0.2 kGy). There were no significant TL differences in the dose dependency among all integration areas for each natural mineral sample. The dose response curves overlapped within the standard deviations. As shown in Figure 5a, there

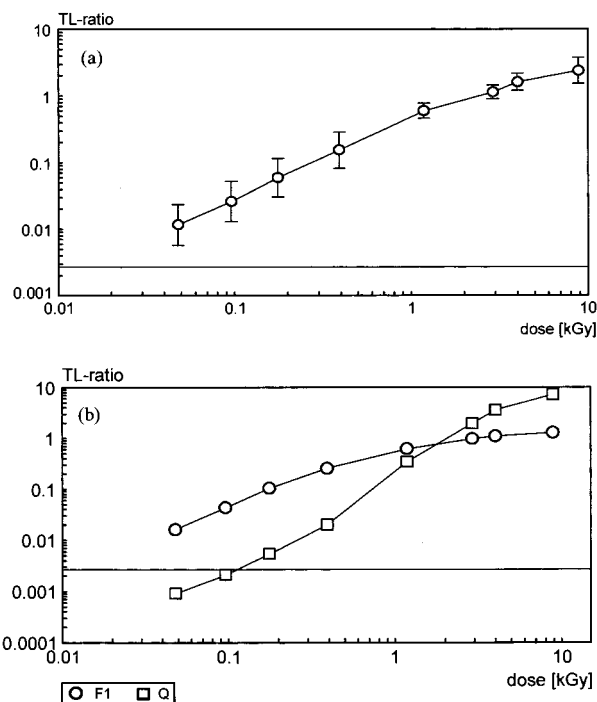


Figure 5. TL dose response of natural mineral samples (a) and standard samples (b) (F1, standard feldspar F1; Q, Merck quartz) in the integration area 200–300 °C. Horizontal lines indicate the detection limit in this integration area, determined by the measurements of three disks of each natural mineral sample (mean plus 3 times the standard deviation) with the same reirradiation procedure.

was a small but not complete saturation behavior above 1 kGy. From analysis of the standard samples, it became clear that there were differences in the dose response of feldspar and quartz. Whereas the quartz samples yielded a line with two turning points, the feldspar dose response begins to deviate from linearity at ~ 0.3 kGy and appears to saturate above 1–2 kGy (Figure 5b). Both responses contribute to the behavior of natural samples described above.

From a comparison of the results of the irradiated natural samples to the detection limits for this reirradiation procedure, it became clear that the lowest doses are detectable in the integration area of 200–300 °C where all mineral samples irradiated by 0.05 kGy could be distinguished from the unirradiated ones. Only the standard Merck quartz needed a dose of 0.2 kGy to exceed clearly the detection limit because of the lower sensitivity. Higher detectable doses were obtained for the three other integration areas (0.2 kGy for the area 70–200 °C, 0.5 kGy for 300–400 °C, and 1 kGy for 400–500 °C).

The mass-normalized intensities of the ESR lines showed a dose dependency different from that of the TL signals. Logarithmic dose response was observed with a distinct saturation behavior at doses below 1 kGy. The line with $g = 1.9975$ had a lower slope than the line with $g = 2.0006$, indicating that these signals had been produced by different paramagnetic species.

Changes of TL Sensitivity by Prior Heating and Irradiation Procedures. These experiments were carried out to investigate the variability of the TL sensitivity by prior heating and irradiation procedures. It was assumed that heating and irradiation can produce new or reduce existing defects in the crystal lattice of a sample, which then serve as trapping centers

during the following irradiation, resulting in a changed sample sensitivity. This would result in different TL intensities emitted by the same sample after different irradiations with the same dose. It is the consequence for routine food control that the determination of the exact dose, used for food treatment, would be influenced by the reproduction method.

By using the reproduction method, the same portion of material is measured after successive irradiation with different doses. The originally applied dose is calculated by extrapolation of the TL intensities obtained. In archaeological dating, the additive dose determination method is used to avoid influence of the change of the sample sensitivity (Aitken, 1985). The availability of several identical aliquots of the minerals is a prerequisite for this method. In food analyses, the procedure of isolating the minerals from the surface of the products commonly does not yield enough material to obtain equal aliquots. Therefore, the additive dose technique is not suitable for food control.

First, investigations were performed to estimate how much the measuring procedure itself influenced the sample sensitivity. The measuring process used included a first heating from ambient temperature to 500 °C within 60 s to record the TL signal. It was followed by a second identical heating to eliminate the background signal by subtraction. In this experiment, all mineral and standard samples were successively irradiated with a low dose (strontium-90 source, 50 Gy) and measured immediately afterward. In this case, the influence of the irradiation was negligible. It was observed that the samples changed their TL sensitivities to different extents. The sensitivity of the standard feldspar F1 in the different integration areas decreased to values of between 93 and 72% as compared to the initial intensity (100%) within 15 successive irradiation and measurement cycles. On the other hand, the pure Amberger feldspar showed a small increase in sensitivity of up to 110%. More rapid changes were observed in the quartz and sea sand samples. There were increases of 150–400% within 15 measurements depending on the integration areas. The Merck quartz sample yielded more drastic changes in the integration range of 400–500 °C, whereas the highest increase of sensitivity in the Merck sea sand occurred in the integration area of 70–200 °C. The natural mineral samples showed sensitivity changes within this variability of the standard samples. Especially the samples consisting almost completely of quartz as indicated by the X-ray diffraction spectra revealed high increases in TL sensitivity.

The samples showed quite different changes in sensitivity caused by extended heating periods (see Materials and Methods), but in general a significant increase in sensitivity due to heating above temperatures of ~350 °C was observed. The lower the temperature of the integration area, the higher the increase that occurred (Figure 6a). The most drastic changes were produced by the pure quartz samples and the natural minerals with high contents of quartz.

In Figure 6b, the results obtained after radiation treatments with different doses and dose rates are shown. Enormous differences between the behaviors of the single samples were observed in this experiment, so that Figure 6b can only demonstrate the general trend but no single results. Doses below 1 kGy did not significantly affect the TL sensitivity of all samples

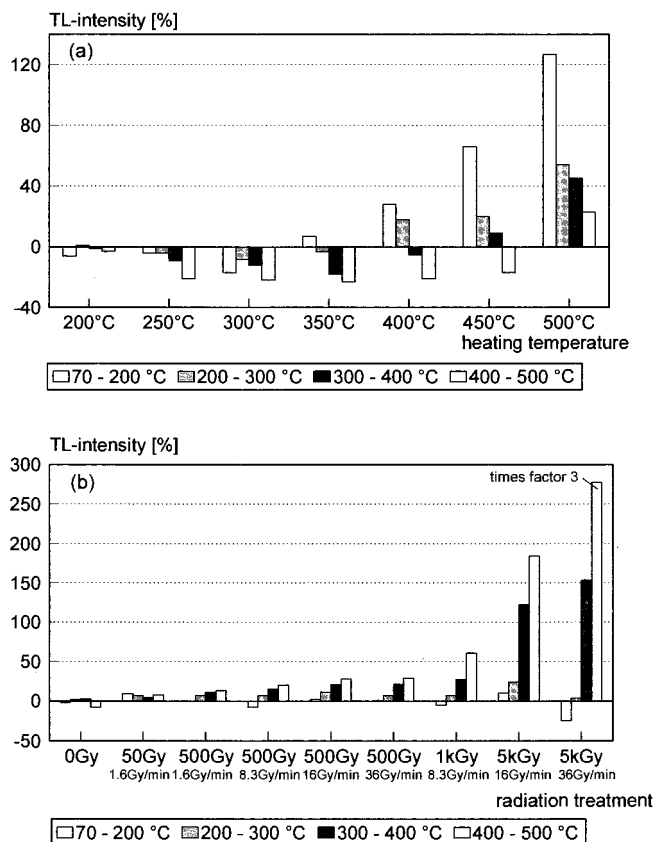


Figure 6. Change of TL sensitivity by heat (a) and radiation (b) treatment: means of 12 natural samples and the 4 standard minerals.

when different dose rates were applied. Irradiation by 5 kGy increased the signal intensities, especially in the integration areas of 300–400 and 400–500 °C. Although the standard feldspars did not reveal this behavior, the sensitivity of the standard quartz samples increased by up to 160 times (Merck quartz, integration area 400–500 °C, 5 kGy, 36 Gy/min) the sensitivity before irradiation with a distinct trend toward the higher dose rate. The most drastic increase of sensitivity was observed for the natural minerals containing a high share of quartz.

In summary, it can be concluded that prior heating and radiation treatment as well as the measuring process itself may significantly influence the TL sensitivity of a sample. The sensitivities of natural samples with a quartz-specific TL behavior are more variable than samples with a TL being more dependent on the contained feldspars. Although the signals in the higher integration areas (300–400 and 400–500 °C) are affected most, also the integration area 200–300 °C, which is most relevant in food routine control, revealed changes in the sensitivities. This might be a reason why there are problems in estimating the applied doses of commercial samples.

CONCLUSIONS

The experiments described have shown that TL analysis is a very sensitive method for detecting food irradiation. Principally, the origin of the minerals isolated from the products does not determine the signal intensities and glow curve shapes. All of the samples investigated that originated from agriculturally used areas or from beaches contained quartz as a major

component and had a more or less minor content of feldspars. Experiments on pure quartz and feldspar samples have shown that feldspars have a much higher sensitivity than the quartz contained in the samples, resulting in a TL behavior of the natural samples that was influenced by both materials. Some principal differences were observed in signal stability, especially concerning photosensitivity, dose dependency, and changes of sensitivity by prior heating and irradiation. A comparison with the irradiation-specific ESR signals has led to the conclusion that TL is not due to the paramagnetic species which are detected by ESR measurements. Because of the existence of high ESR signals in the natural samples, which had not been artificially irradiated, and the much higher amounts of mineral material, being necessary for ESR analysis, this technique is considered to be unsuitable for the detection of irradiated foods by using minerals isolated from the surface.

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LITERATURE CITED

- Aitken, M. J. *Thermoluminescence Dating*; Academic Press: London, 1985; p 19.
- Autio, T.; Pinnioja, S. Identification of irradiated foods by the thermoluminescence of mineral contamination. *Z. Lebensm. Unters. Forsch.* **1990**, *191*, 177–180.
- Autio, T.; Pinnioja, S. Identification of irradiated foods by the thermoluminescence of the contaminating minerals. In *Recent Advances on the Detection of Irradiated Food*; BCR Information, EUR/14315/EN; Leonardi, M., Raffi, J. J., Belliardo, J. J., Eds.; Commission of the European Communities: Brussels, 1993; pp 177–185.
- Bayliss, P., Berry, L. G., Mrose, M. E., Smith, D. K., Eds. *Mineral Powder Diffraction File. Data Book*; JCPDS International Centre for Diffraction Data: 1980.
- Bøtter-Jensen, L. The automated Risø TL dating reader system. *Nucl. Tracks Radiat. Meas.* **1988**, *14*, 177–180.
- EN 1788. *Foodstuffs—Detection of irradiated food from which silicate minerals can be isolated—Method by thermoluminescence*; European Committee for Standardization (CEN) Secretariat: Brussels, 1997.
- Heide, L.; Bögl, K. W. *Die Messung der Thermolumineszenz—Ein neues Verfahren zur Identifizierung strahlenbehandelter Gewürze*; Bericht des Instituts für Strahlenhygiene des bundesgesundheitsamtes: Neuherberg, Germany, 1984; ISH-Heft 53.
- Mahesh, K.; Wenig, P. S.; Furetta, C. *Thermoluminescence in Solids and Its Applications*; Nuclear Technology Publishing: Ashford, Kent, England, 1989; p 98.
- McKeever, S. W. S. *Thermoluminescence of Solids*; Cambridge University Press: Cambridge, U.K., 1988; p 176.
- Sanderson, D. C. W.; Slater, C.; Cairns, K. J. Thermoluminescence of foods: Origins and implications for detecting irradiation. *Radiat. Phys. Chem.* **1989a**, *34*, 915–924.
- Sanderson, D. C. W.; Slater, C.; Cairns, K. J. Detection of irradiated food. *Nature* **1989b**, *340*, 23–24.
- Sanderson, D. C. W.; Carmichael, L. A.; Clark, P. A.; Clark, R. J. *Identification of Irradiated Fruits and Vegetables*; Development of luminescence tests to identify irradiated foods, Final Report N1701; MAFF N1701: London, Great Britain, 1992.
- Sanderson, D. C. W.; Carmichael, L. A.; Naylor, J. D. Recent advances in thermoluminescence and photostimulated luminescence detection methods for irradiated foods. In *Detection Methods for Irradiated Foods—Current Status*; McMurray, C. H., Stewart, E. M., Gray, R., Pearce, J., Eds.; Royal Society of Chemistry: Cambridge, U.K., 1996; pp 124–138.
- Schreiber, G. A.; Ziegelmann, B.; Quitzsch, G.; Helle, N.; Bögl, K. W. Luminescence techniques to identify the treatment of foods by ionizing radiation. *Food Struct.* **1993a**, *12*, 385–396.
- Schreiber, G. A.; Wagner, U.; Helle, N.; Ammon, J.; Buchholtz, H.-V.; Delincée, H.; Estendorfer, S.; von Grabowski, H.-U.; Kruspe, W.; Mainczyk, K.; Münz, H.; Schleich, C.; Vreden, N.; Wiezorek, C.; Bögl, K. W. Thermoluminescence analysis to detect irradiated fruit and vegetables—an intercomparison study. Bericht des Instituts für Sozialmedizin und Epidemiologie des Bundesgesundheitsamtes: Berlin, 1993b; SozEp-Heft 3/1993.
- Schreiber, G. A.; Hoffmann, A.; Helle, N.; Bögl, K. W. Methods for routine control of irradiated food: determination of the irradiation status of shellfish by thermoluminescence analysis. *Radiat. Phys. Chem.* **1994**, *43*, 533–544.
- Schreiber, G. A.; Helle, N.; Bögl, K. W. An inter-laboratory trial on the identification of irradiated spices, herbs and spice-and-herb mixtures by thermoluminescence analysis. *J. AOAC Int.* **1995**, *78*, 88–93.

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